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AN INVESTIGATION OF METHODS OF USING A GRAPHITE RESISTANCE HEATER WITH AIR

VAS

BOGDONOFF

PRINCETON UNIVERSITY
PRINCETON, NEW JERSEY

OCTOBER 1963

AEROSPACE RESEARCH LABORATORIES
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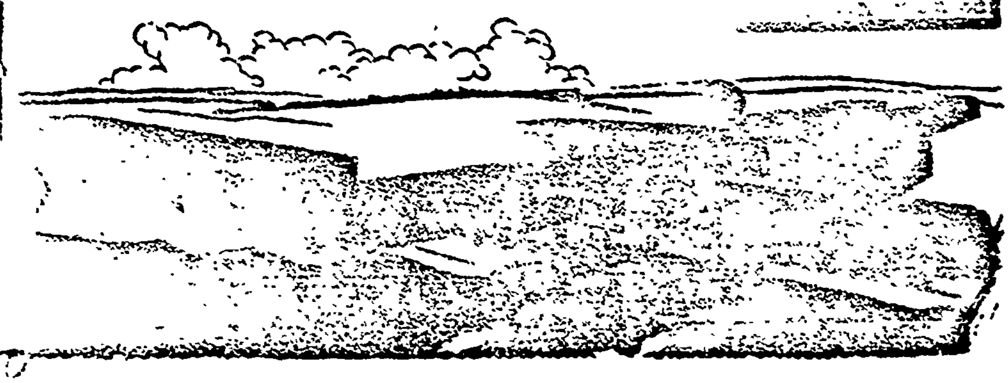

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AN INVESTIGATION OF METHODS OF USING A GRAPHITE RESISTANCE HEATER WITH AIR

10 by I. E. VAS and
S. M. BOGDONOFF,

PRINCETON UNIVERSITY
PRINCETON, NEW JERSEY

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17 Task 7065-01

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AEROSPACE RESEARCH LABORATORIES
OFFICE OF AEROSPACE RESEARCH
UNITED STATES AIR FORCE
WRIGHT-PATTERSON AIR FORCE BASE, OHIO

FOREWORD

This final technical report was prepared by the Gas Dynamics Laboratory, Princeton University, Princeton, New Jersey, on Contract AF 33(657)-8818 for the Aerospace Research Laboratories, Office of Aerospace Research, United States Air Force. The work reported herein was accomplished on Task 7065-01, "Fluid Dynamics Facilities Research" of Project 7065, "Aerospace Simulation Techniques Research" under the technical cognizance of Mr. Emil J. Walk of the Fluid Dynamics Facilities Laboratory of ARL.

ABSTRACT

An investigation of the possibility of using a graphite heater as a basis for a air heating device SHOWED ~~has shown~~ that the three most promising materials to coat the graphite, boron nitride, hafnium carbide, and zirconium carbide, were not satisfactory. Other methods for heating air are suggested.

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INTRODUCTION

For the past three years, the Gas Dynamics Laboratory of Princeton University, has been developing and using graphite resistance heaters with nitrogen, to obtain the high temperatures at high pressures required to drive hypersonic wind tunnels. The basic ideas and varied developments of this heater are detailed in references 1 - 4. The concept of the heater is quite simple. If a non-oxidizing gas is to be heated, several materials have the ability of withstanding the high temperature and, at the same time, have characteristics which permit easy resistance heating. Graphite is one such material, and the heater designs have been concerned with the detailed geometry and material of which these heaters have been built. Specific attention has been placed on the development of a graphite heater for nitrogen (which closely simulates air in it's fluid mechanical characteristics). The early heaters were made of a type of graphite known as ATJ and, because of the porosity and impurity problems associated with this type of graphite, were provided with a pyrolytic vapor deposited graphite coating. The graphite element of the general geometry shown in Figure 1, with such pyrolytic coatings, proved quite satisfactory for performance up to about 1000 lbs. per square inch and temperatures of the order of 5000° Fahrenheit.

In the last year or two however, there have been considerable improvements in the types of graphite available. The newer specimens were denser and purer. In the configurations now in use, ZTA graphite is used directly with no coatings and has provided very satisfactory operation at pressures of

5000 lbs. per square inch and temperatures as high as 5700° Fahrenheit. The heaters provide operation at these elevated temperatures and pressures for many hours with failures primarily traceable to either very small impurities in the outer shell, which slowly sublimes and permits leakage of the gas to by-pass the heater, or to mechanical handling of the heater during inspection periods. The heaters have been quite adequate for high Mach number testing in a range not heretofor available in continuous facilities, with non-contaminated gas streams.

PURPOSE AND SCOPE OF PRESENT STUDY

The main purpose of the subject study was an attempt to extend the techniques outlined above to heating air with it's problems of oxidation. The graphite heater can not be used directly, since at the temperatures desired, graphite would burn in air. The first part of the present program was a study of the feasibility of two methods which had been conceived during the work on the graphite-nitrogen development.

OXIDATION RESISTANT COATINGS FOR GRAPHITE

One approach was an attempt to use the basic graphite heater as the key element, but to protect the graphite from the oxygen in the air. This approach had several inherent advantages. The graphite itself is cheap, easy to fabricate, and has excellent electrical characteristics for a resistance heater. Graphite at high temperature is relatively strong and shock resistant. At the same time, there are rather major programs in the country devoted to the materials problems of protecting graphite from oxidation in

rocket nozzles. There have been several methods of applying such coatings which have good mechanical strength and, on the basis of the rocket tests, were able to protect the graphite under very severe conditions.

The application of this information and technique is not straightforward. The rocket development work, and also some work which has been associated with the protection of nose cones and wing leading edges, is a problem in which the heat input is into the graphite. The protective coating is exposed to very high temperature, while the graphite is cooled by either its heat capacity or other cooling techniques. In the application proposed herein, the heating is from the graphite through the protective coating to the air, that is, the heat flows in the opposite direction, and the temperatures within the graphite may very well be considerably higher then that in the protective coatings. A study of the materials which have been used to protect graphite and their methods of application seemed to indicate some considerable possibility of success in the fabrication and use of such coatings for the graphite heater and appear to present no inherent difficulties with the following exceptions: The coatings have been applied to rocket nozzles of rather simple shape and large dimensions. The present application is to a rather small, considerably more complicated geometry, which involves enclosed passages. Second, although rather crude overall tests have been made of the materials used to protect graphite, there is no direct data on the conditions of temperature and pressure and desired long life, which were

the requirements for our present heater design.

TWO STAGE HEATER

The second concept of heater design tried to simplify the problem by separating the heating and the oxidation resistant functions of the heater. Graphite in an inert atmosphere had already been well proven as an excellent method for heating. At the same time, an examination of the radiation from the graphite heater at these very high temperatures, indicated that this in itself might be an excellent heating source. A concept was therefore developed of using graphite in an inert atmosphere to heat by radiation a ceramic tube through which the air passed. In such a system, schematically outlined in Figure 2, there would be no contact between the oxygen and the graphite and many of the problems associated with specifying heater material characteristics with ceramic could be eliminated. The ceramic tube would be required to act only as a barrier between nitrogen and oxygen in the air in the tube. This could be done by adjusting the pressure through the tube and the inert atmosphere to be about equal. There is no difficulty in controlling the rate of heating from the graphite heater by programing the electrical input, so that heat shock would not be a problem. Although this method holds considerable promise, the detailed design revealed that there would be considerable mechanical complexity in sealing and arranging for differences in expansion coefficients, pressure balancing, etc.

DETAILED FEASIBILITY STUDIES

In the first approach, that of coating the graphite element, there is a considerable body of information already available on coating materials from experimental results on rocket nozzles. A study of these methods and techniques yielded three materials which appeared to be particularly promising, boron nitride, hafnium carbide, and zirconium carbide, with coating techniques which were either of the spray type or diffusion deposited. The boron nitride is machinable, but the carbides are quite difficult to machine and therefore, for the latter two, coating of the completely assembled element is required.

In the second approach, the two stage heater design, the examination of the problem showed some problems beyond that of the multiple seals, etc. All of the ceramics which were examined for use in the present study, were, to some degree, porous, this resulted in the possibility that some of the air and it's attendant oxygen, might contaminate the inert gas around the graphite heater. Such a phenomena would immediately cause oxidation and distruction of the graphite heater. The only way to solve this problem was to require that the pressure of the inert gas be slightly above that of the air in the ceramic tube so that the porosity of the ceramic would result in a small leakage of the inert gas into the test gas. This would assure that no oxygen came in contact with the graphite heater. This type of control had to be quite sensitive since large pressure differences between the inert gas and the air would result in

considerable contamination of the air sample with the inert gas. The use of nitrogen as an inert gas would have minimum effects due to contamination. The second part of the study was the question of trying to eliminate the gas flow system in the outer radiation heater made of graphite. It appears however, that this gas flow is necessary to keep the temperature of the outer heater uniform. It has been shown by other investigators, that graphite cylinders used as resistance heaters for high current applications, suffered from "channeling". This appears as though the high current prefers a path down one side of the cylinder, and heating was not uniform. The use of the spiral passages in the present heater, and it's attendant nitrogen flow, appears to be the key factor in uniformly distributing the heat around the entire graphite cylinder.

On the basis of these feasibility studies, the decision was made to concentrate on the coating technique as the method most likely to give a practical solution in a reasonable time scale. As a result, the somewhat more complicated two stage system was not studied further.

COATING PROGRAM AND RESULTS

The major effort on materials and the direct application of the coatings was carried out primarily by High Temperature Materials of Cambridge, Mass. Their background, experience, and cooperation, were the main support of a current program in it's attempt to obtain a satisfactory solution to the oxidation resistance coated graphite problem.

The first approach was to use pyrolitic boron nitride, deposited on graphite. The heater elements, similar in overall design to the previous spiral shell type, were made somewhat smaller in dimension so that a plating thickness of about .020 inches of boron nitride could be deposited on the basic graphite core. The shell and the spindle were to be coated separately, and then finally machined, fitted and assembled. Although boron nitride had been used quite widely in many other applications, several questions as to it's suitability for the present work had to be investigated. The erosion rate at high pressures were completely unknown. Data was available only at a few atmospheres pressure as compared to the 1000 lb. per square inch and higher as desired for the present heater design. At the same time, the interaction of boron nitride with graphite at these conditions was unknown. A study of these two phenomenon resulted in information that a chemical reaction takes place between boron nitride and carbon in air, in which oxides of boron are formed. This oxide is a liquid under 2000^{o} Centigrade as compared to the considerably higher liquification and evaporation points of the graphite and the boron nitride. As a result of this study, the boron nitride coating program was dropped.

The second approach was to use a coating of hafnium carbide. Considerably more was known of the hafnium carbide and it exhibited several characteristics which appeared to be quite promising. It had very good erosion resistance and it was also electrically conductive. Tests with a plasma torch at atmospheric

pressure indicated it's ability to withstand quite high temperatures, and it was possible to apply very thin coatings by several techniques. Diffusion coatings seemed to be particularly good. The key unknown was the erosion resistance, but several claims had been made as to it's possible application in this area. One additional difficulty was the problem of machining this material. In it's final form, it is almost unworkable by standard machine techniques. As a result, the possible application for the hafnium carbide had to be carried out with the heater element in one piece, that is, the spindle and shell fully assembled. While this study was underway, a suggestion from High Temperature Materials that a mixed zirconium carbide, hafnium carbide coating, would probably have better characteristics, resulted in our dropping further work on the pure hafnium carbide coating.

At high temperature, stable oxides of zirconium and hafnium are formed in the suggested combination, with the possibility of being able to reach temperatures of the order of 2700° Centigrade. Tests of the oxides and the carbides showed very good characteristics. The oxides are insulators, but the carbides are conductors. The key problem associated with this approach was the question of thermal expansion of the coatings. The expansion of the carbides is higher than that of the graphite and no information was available as to the mechanical strength or stability of coatings with these significant differences in the thermal expansions. A diffusion coating process was chosen and a basic program on the diffusion coating of this mixed carbide on three different types of graphite was undertaken. The graphites used were ATJ, ZTA and CD05 (all from the

National Carbon Company). ATJ was one of the original graphites used in the graphite heater. This material has some porosity but mechanical characteristics which appear to be advantageous for the present application. ZTA graphite, which is more dense than the ATJ graphite, posed the question as to whether sufficient bonding of the diffusion coating in the graphite would take place. Type0005 graphite fell between ATJ and ZTA in certain mechanical characteristics. Three sets of spindles and shells (Figure 3) were made of the different types of graphite with the design modified so that plating of about .020 inches of the zirconium-hafnium carbide combination would give the final dimensions desired.

Although a rather considerable effort was involved in attempting to successfully coat these elements, it was found that the vapor deposition technique could not be made to work satisfactorily on the configuration desired. Additional study also showed that the oxidation of the zirconium-hafnium carbide would not stabilize, and that as time went on at the elevated temperatures, all of the carbide would be converted and the graphite would be exposed directly to the hot oxides, a condition which would result in direct oxidation of the graphite.

CONCLUDING REMARKS

As a result of the above detailed studies, no satisfactory coating for the graphite resistant element was found, and at the moment, there are no materials or techniques which appear to hold promise greater than those which were used in

the original program. It should be pointed out however, that no positive statement of this sort can be supported because of the lack of direct information on the characteristics of many of these materials under the desired conditions.

The carbides looked promising and perhaps in solid form, that is without the graphite core, might have some possibility as direct heating units. Further detailed examination of their electrical propertities with temperature are required.

The small geometrical scale of the present heater design, made the coatings particularly difficult and there is some possibility that larger scaled tests may take care of this particular difficulty in the present program. However, it should be pointed out that the basic limitations, although influenced by the depositing technique, were primarily the chemical reactions and not the scale of the study.

In the light of the failure of the coating program to achieve any success, any future work should probably concentrate on the two stage design, even with it's mechanical complexity. Another type of two stage heater might also be reasonable. Using the present graphite heater to heat nitrogen, oxygen from a separate heater at a lower temperature, might be mixed to reconstitute the air in a small settling chamber ahead of the nozzle. The potential of this system results in a somewhat lower temperature than the straight graphite-nitrogen design now in use because of the limitation in heating the pure oxygen. It also suffers from the requirement that pure nitrogen be still used in the major part of the heater, and the simplicity of using air

directly to drive a hypersonic tunnel is lost. The system will also be complex because of the necessity of mixing the gases in the small settling chamber at high pressure and temperature, and the necessity for very careful metering devices, so that the constitution of the final test gas is known.

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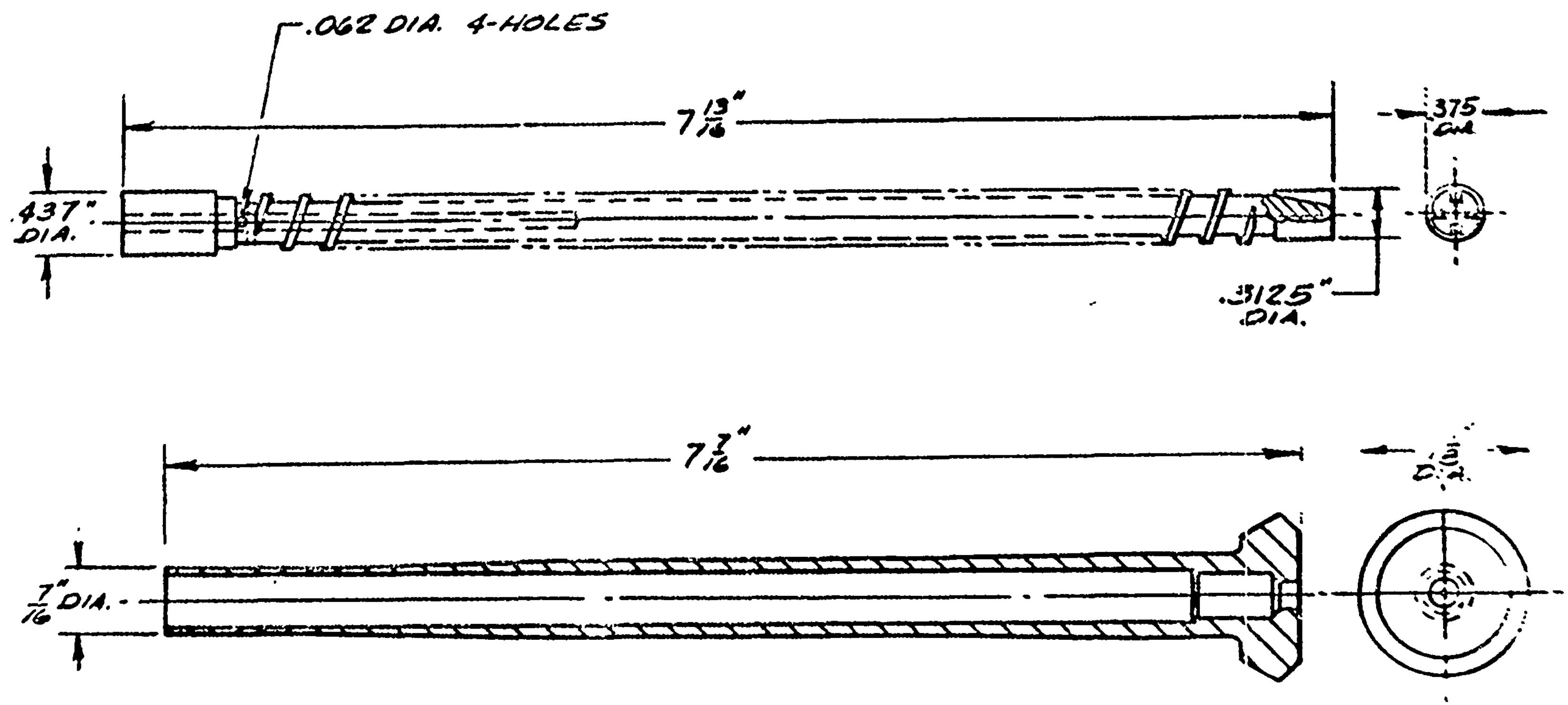

Figure 1. Sketch of Heater Element

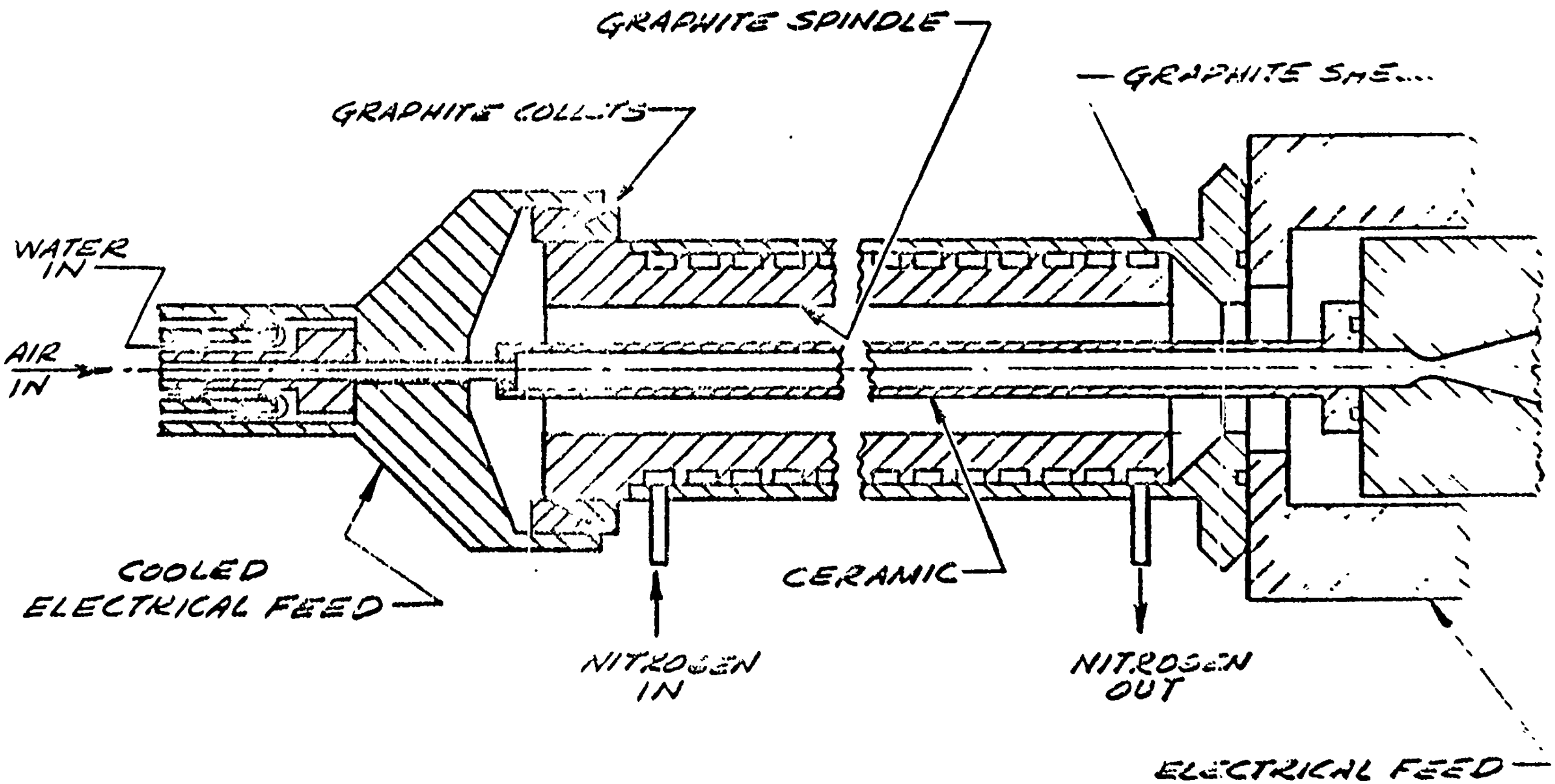

Figure 2. Sketch of Ceramic Heater with Graphite Radiation Heater

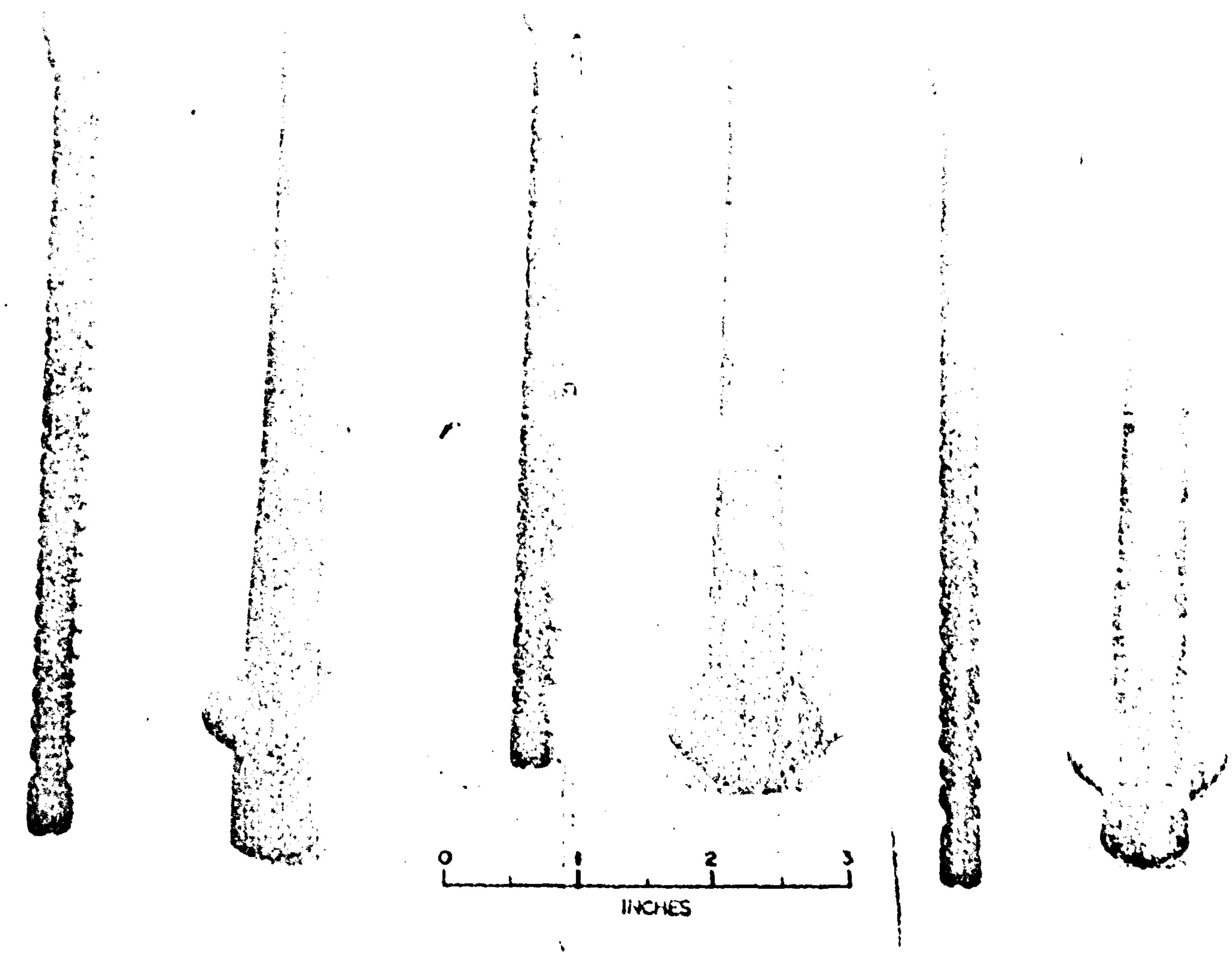

Figure 3. Photograph of Spindle and Shell for Cooling Purposes